Collective excitations and the nature of Mott transition in undoped gapped graphene

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Particle-hole continuum (PHC) for massive Dirac fermions in presence of short range interactions, provides an unprecedented opportunity for formation of two collective split-off states, one in the singlet and the other in the triplet (spin-1) channel in undoped system. Both poles are close in energy and are separated from the continuum of free particle-hole excitations by an energy scale of the order of gap parameter Δ. They both disperse linearly with two different velocities reminiscent of spin-charge separation in Luttinger liquids. When the strength of Hubbard interactions is stronger than a critical value, the velocity of singlet excitation which we interpret as a charge boson composite becomes zero, and renders the system a Mott insulator. Beyond this critical point, the low-energy sector is left with a linearly dispersing triplet mode – a characteristic of a Mott insulator. The velocity of triplet mode at the Mott criticality is twice the velocity of underlying Dirac fermions. The phase transition line in the space of U and Δ is in qualitative agreement with a more involved dynamical mean field theory (DMFT) calculation.

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INTRODUCTION

Graphene is a huge network of sp^2 bonds formed between carbon atoms on a two-dimensional (2D) sheet [1]. The low-energy effective model for p_z electrons in this system is the 2+1 dimensional Dirac theory which contains an energy scale proportional to v_F – the Fermi velocity of electrons [2]. Effects of interactions and/or disorder can be taken into account on top of this non-interacting fixed point [2]. Breaking the sub-lattice symmetry gives rise to a gap in the single-particle excitations spectrum [3], which introduces the gap parameter Δ as another energy scale. The gapped graphene can be modeled with 2+1 dimensional massive Dirac fermions [4]. Gapped graphene supports interesting new class of excitations in the spectrum such as, domain walls [5]. Moreover the single-particle gap (mass) gives rise to suppression of Coulomb fields at nano-meter length scales [6]. The gap could also give rise to universal nonlinear optical response [7]. When the many-body interactions of various form are added to the massive Dirac theory, the nature of many-body excitations becomes even more interesting. Recent *ab-initio* estimates of the strength of the Coulomb repulsion in various forms of graphene suggests that the Hubbard parameter in graphene can be quite remarkable [8], which introduces a third energy scale, U. Therefore, an interesting question here would be the effects of local Hubbard type interactions on the excitation spectrum of gapped graphene, and the nature of transition to Mott insulating phase in gapped graphene. Our recent full-fledged DMFT investigation of the so called ionic-Hubbard model on the honeycomb lattice suggested three phases [9]: (i) Band insulating phase for $\Delta \gg U$. (ii) Mott insulating phase for $\Delta \ll U$, and (iii) a semi-metallic phase for $U \sim \Delta$. The above study is about the nature of ground state. In order to study the connection of excited states and the ground state, one notes that; deep in the Mott insulating phase, the energy scale U is expected to be dominant over Δ, such that the low-energy excitations are expected to be spin fluctuations arising from super-exchange interaction induced by the large U. We would like to focus on, the evolution of the collective spin and charge dynamics in this system as a function of the Hubbard U for a system with a non-zero gap parameter Δ. It turns out that the presence of a non-zero gap parameter facilitates the separation of two collective states each of which has a different velocity, reminiscent of the spin-charge separation in Luttinger liquids.

FORMULATION OF THE PROBLEM

The Hamiltonian we consider here is the so called ionic-Hubbard model, which is defined by

$$H = \hbar v_F \sum_{\mathbf{k}s} \psi^\dagger_{\mathbf{k}s} \left[\sigma.\mathbf{k} + \sigma_z \Delta\right] \psi_{\mathbf{k}s} + U \sum_j (n^a_{j\uparrow} n^a_{j\downarrow} + n^b_{j\uparrow} n^b_{j\downarrow}) \tag{1}$$

where $n^f_{js} = f^\dagger_{js} f_{js}$ with $f = a, b$ corresponding to number operator at the j'th unit cell at sub-lattices, A, B, respectively. The spinor notation $\psi^\dagger_{\mathbf{k}s} = (a^\dagger_{\mathbf{k}s}\ b^\dagger_{\mathbf{k}s})$ has been used. Here $s =\uparrow,\downarrow$ denotes the z component of the physical spin, $\mathbf{k} = (k_x, k_y)$ is a two-dimensional momentum vector, and σ stands for Pauli matrices. U is the strength of the on-site Coulomb interaction known as Hubbard parameter, and Δ is the mass parameter. The quadratic part of this Hamiltonian can be diagonalized by a simple unitary transformation to the basis of conduction $(+)$ and valence $(-)$ states, and the corresponding eigen-values are given by,

$$\varepsilon^\pm(\mathbf{k}) = \pm v_F \sqrt{|\hbar\mathbf{k}|^2 + \Delta^2}, \qquad \Delta \neq 0, \tag{2}$$

where $\hbar v_F = \sqrt{3} ta/2$ and a being the C-C bond length. The hopping amplitude between the nearest neighboring carbon atoms is $t \sim 2.8$ eV. Due to paramagnetic nature of the non-interacting system (i.e. $U = 0$), the occupation numbers and energies of the conduction and valence bands are independent of spin orientation. We consider the undoped system with pre-

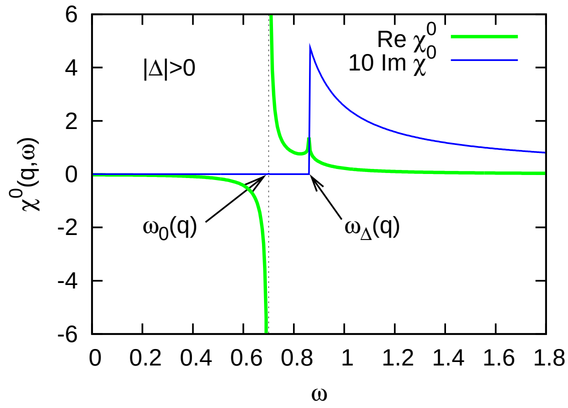

FIG. 1. (Color online) Behavior of free particle hole propagator $\chi^0(\mathbf{q},\omega)$. The imaginary part has been magnified to clearly mark the onset of particle-hole continuum of free particle-hole excitations. In this figure, vertical scales are drawn for $qa = 0.7$, and v_F and $\hbar$ are assumed to be unit.

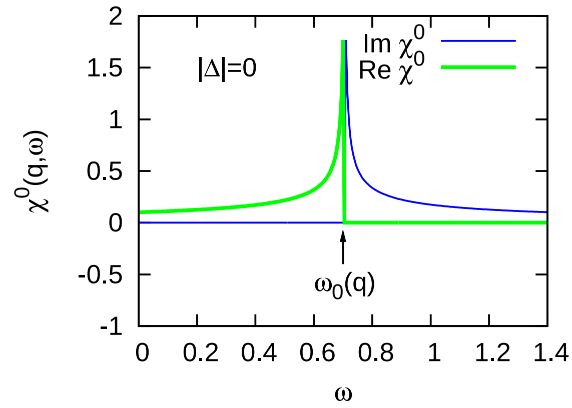

FIG. 2. (Color online) Behavior of free particle hole propagator $\chi^0(\mathbf{q},\omega)$ when the gap vanishes, i.e. $\Delta = 0$. The imaginary part (blue line) marks the onset of particle-hole continuum of free particle-hole excitations. In this figure, vertical scales are drawn for $qa = 0.7$, and v_F and $\hbar$ are assumed to be unit.

cisely one p_z electron per carbon atom, and assume the temperature to be zero. Then we extend the collective mode analysis of Ref. [11] to the case with $\Delta \neq 0$, whereby the single-particle spectrum changes from $\pm\hbar v_F|\mathbf{k}|$ to the one given by Eq. (2). This change in the single-particle spectrum, changes the borders of the particle-hole continuum. However, as will be show here, at the border corresponding to massless spectrum, interesting collective quanta can be made in presence of short range Coulomb interactions.

Using equation of motion it can be shown that a simple RPA-like expression governs the eigen-value equation for the collective excitations in the triplet and singlet channels of the two-band arising from Dirac fermions in presence of the short range Coulomb interactions are given by,

$$1 \pm U\chi^0(\mathbf{q},\omega) = 0, \tag{3}$$

where the $-$ $(+)$ sign corresponds to triplet (singlet) channel [12], and the sign difference can be traced back to fermion anti-commutation relation. Note that although in the above expression, χ^0 is the particle-hole (polarization) bubble, the (approximate) bosonic operators which satisfy the above equation are of a peculiar form, which *does not* necessarily coincide with the precise particle-hole fluctuation form [11]. The non-interacting susceptibility χ^0 employed in the above equation, is defined by,

$$\chi^0(\mathbf{q},\omega) = \frac{1}{N}\sum_{\mathbf{k}} \frac{\bar{n}^+_{\mathbf{k}+\mathbf{q}} - \bar{n}^-_{\mathbf{k}}}{\omega + i\eta - (\varepsilon^+_{\mathbf{k}+\mathbf{q}} - \varepsilon^-_{\mathbf{k}})}. \tag{4}$$

For the dispersion relation (2), the susceptibility has been calculated by many authors, the analytic form of which reads [13, 14]:

$$\chi^{(0)}(\mathbf{q},\omega) = -\frac{|\mathbf{q}|^2}{\pi\hbar(v_F^2|\mathbf{q}|^2-\omega^2)}\left\{\frac{2\Delta}{\hbar} + \frac{v_F^2|\mathbf{q}|^2-\omega^2-4\Delta^2/\hbar^2}{\sqrt{v_F^2|\mathbf{q}|^2-\omega^2}} \arctan\frac{\sqrt{v_F^2|\mathbf{q}|^2-\omega^2}}{2\Delta/\hbar}\right\}. \tag{5}$$

To understand the structure of this function, in Fig. 1 we have plotted the real and imaginary part of this function versus ω for some representative value of $\mathbf{q}$. As can be seen, the imaginary part is non-zero for $\omega > \omega_\Delta(\mathbf{q})$, where

$$\omega_\Delta(\mathbf{q}) = \sqrt{v_F^2|\mathbf{q}|^2 + 4\Delta^2/\hbar^2}, \tag{6}$$

corresponds to the lower boundary of PHC of massive Dirac fermions. The singularity of the above inter-band response at $\omega = \omega_\Delta(\mathbf{q})$ is of a weak logarithmic form which could give rise to a solution for Eq. (3) only in the triplet channel, and for extremely large values of U which is unphysical. The χ^0 however possesses another much more interesting simple pole structure at a lower energy scale, $\omega = \omega_0(\mathbf{q})$ given by,

$$\omega_0(\mathbf{q}) = v_F|\mathbf{q}|. \tag{7}$$

Note that this equation defines a line in the plane of ω and $|\mathbf{q}|$, which corresponds to the lower boundary of the PHC of massless ($\Delta = 0$) Dirac fermions and is determined by the characteristic velocity, v_F of the underlying Dirac fermions. This could be considered as a property of a "parent" massless Dirac system which manifests itself as a simple-pole structure when a mass parameter, Δ is turned on in the non-interacting part of the Hamiltonian. At the gapless point, where $\Delta = 0$, the two energy scales at $\omega_0(\mathbf{q})$ and $\omega_\Delta(\mathbf{q})$ merge and the resulting singularity of χ^0 will be of inverse-square-root form [10].

As can be seen, to search for solutions of Eq. (3), one has to look for the intersection of the real part (green line) with

the constant horizontal line $\mp\frac{1}{U}$, where the upper (lower) sign corresponds to singlet (triplet) channel. As can be very clearly seen in the figure, near $\omega_0(\mathbf{q}) = v_F|\mathbf{q}|$, where the imaginary part of χ^0 is zero, there can be poles both in singlet and triplet channel. The simple pole structure of the particle-hole propagator near this energy scale implies that the poles exist for every value of the Hubbard U, no matter how small or how large it is. To see this more clearly, note that the real part of the susceptibility near $\omega_0(\mathbf{q})$ behaves as,

$$\mathrm{Re}\chi^0(\mathbf{q},\omega) \approx \frac{|\mathbf{q}|\Delta}{\pi v_F \hbar^2 \left[\omega - \omega_0(\mathbf{q})\right]}. \tag{8}$$

The solutions of the eigenvalue equations (3) for arbitrary value of U define two collective branches in singlet and triplet channel as,

$$\omega_{\text{singlet/triplet}}(\mathbf{q}) = v_F q \left(1 \mp \frac{U\Delta a^2}{\pi\hbar^2 v_F^2}\right), \tag{9}$$

where upper sign corresponds to the singlet channel, and the lower sign corresponds to the triplet channel. These solutions are available even for arbitrarily small of U. This formula can be interpreted as the spin charge separation in the sense that the singlet and the spin-1 modes move at different velocities given by,

$$v_{c/s} = v_F \left(1 \mp \frac{U\Delta a^2}{\pi\hbar^2 v_F^2}\right). \tag{10}$$

The mere existence and simple pole structure of the χ^0 at the energy scale ω_0 is a unique consequence of the gap opening in the single-particle spectrum of excitations. The presence of gap in the single particle spectrum, pushes the continuum of free particle-hole pairs to higher energies, and hence the above collective excitations which appear around $\omega_0(\mathbf{q}) = v_F|\mathbf{q}|$ line are well separated from the boundary of PHC and therefore will be protected from Landau damping to the incoherent background of free particle-hole excitations.

DISCUSSION

The opening of a single particle gap in the spectrum of excitations in graphene, although pushes the lower edge of the particle-hole continuum from the energy scale ω_0 to energy scale ω_Δ, but still leaves behind a signature of underlying massless Dirac fermions in the form of a singular behavior for χ^0 on the line defined by $\omega_0(\mathbf{q}) = v_F|\mathbf{q}|$ corresponding to the PHC boundary of the underlying massless Dirac fermions. The simple pole left on this line has two consequences: (i) Due to the sign change of the susceptibility across ω_0, there will be collective mode solutions at both singlet and triplet channels, for arbitrary values of the Hubbard U. The explicit form of the solutions are given by Eq. (9). (ii) The different velocities for the two modes is reminiscent of the situation one encounters in one-dimension, where a faster ”spin mode” overtakes the slower ”charge mode”. Albeit the difference is that in 1D the divergences in particle-hole bubbles are of the characteristic inverse square root type. In the case of 2D massive Dirac fermions, where Δ is finite, the divergence in the particle-hole propagator is of a simple pole form. When the limit $\Delta \to 0$ is taken, the simple pole merges with the logarithmic singularity at the boundary of PHC, ω_Δ, and gives rise again to a inverse-square-root behavior for gapless Dirac fermions in 2D [10]. It is therefore the non-zero value of Δ which provides a solution in the singlet channel in addition to the known solution of the triplet channel at $\Delta = 0$ point [10, 11]. In this sense, it appears that the non-zero value of Δ enhances the separation of the triplet and the singlet modes. Therefore the smallest value of the gap parameter, Δ, totally changes the nature of collective excitations in graphene. This observation may have implications for novel approaches to the bosonization of the 2+1 dimensional massive Dirac fermions. Note that if the interaction employed was of a long-range Coulomb form, the solution in the spin-1 channel would be of a linearly dispersing gapped form, and the solution in the singlet channel would not exist at all. Therefore the two modes discussed here are peculiar feature of short range interactions, which due to remarkable value of the Hubbard U in graphene, may have relevance to physically fabricated samples of gapped graphene.

Now let us discuss the nature of the phase transition marked by vanishing of the velocity of the singlet mode. As can be seen in Eq. (10), when the Hubbard U is large-enough to satisfy,

$$U_c\Delta = \frac{3\pi}{4}t^2, \tag{11}$$

the velocity of singlet excitations becomes zero, and the low-energy sector is exhausted by only triplet excitations of the form

$$\omega_{\mathrm{triplet}}(\mathbf{q}) = 2v_F q. \tag{12}$$

Thinking in the spirit of slave-boson approach [15], an electron (hole) can be assumed to be composed of its spin part, the spinon, and the charge part the doublon (holon). Therefore a composite object constructed from an electron and a hole can be thought of a separate spin-1 composite of two spinons, and a charge composite of a doublon and a holon giving rise to a spin zero boson. Hence the triplet mode can be interpreted as a triplet bound state of two spinons [10, 11], and the new singlet mode the emergence of which is being facilitated by a non-zero gap in the single-particle spectrum, can be interpreted as a composite boson constructed from a doublon and a holon the total charge of which is zero, as it should be. In this sense, the vanishing of the velocity of singlet mode can be associated with infinite enhancement of the effective mass of charge bosons. This gives rise to localization of charge carriers and hence is a Mott transition [16]. Indeed the decreasing trend $U_c \propto \Delta^{-1}$ as a function of the gap parameter Δ in Eq. (11) is in qualitative agreement with our previous

DMFT result for the phase boundary of the Mott insulating phase in the ionic Hubbard model [9]. Note that this agreement holds for very small to moderate values of Δ. For large values of Δ, the phase boundary in DMFT will be given by $U \propto \Delta$ [9]. However, as long as experimental realization of gap parameter in graphene is concerned, Δ will be on the scale of few tens of meV, which is quite small in the scale of the hopping term $t \sim 2.8$ eV. Moreover, the fact that the only low-energy excitations left in the system when U goes beyond U_c is a another evidence that at U_c the systems becomes a Mott insulator. Because in the Mott insulator, the only possible low-energy modes are spin excitations.

When U is further increased, it can be seen in Fig. 1 that the real part of χ^0 and $+\frac{1}{U}$ (triplet channel) will intersect in another point which is immediately below the edge of the PHC, $\omega_\Delta(\mathbf{q})$, and therefore a second triplet mode at slightly higher energy than the first one is expected to form, which is anticipated due to weak logarithmic singularity at ω_Δ. However, this mode is not likely to be realized as it requires quite a large value of U which falls already in the Mott insulating phase, where the ground state maybe totally deformed with respect to the initial starting point of massive Dirac fermions employed in this work.

It is interesting to note that, although the velocity of the underlying Dirac fermions is v_F, the velocity of the spin-1 mode is always more than v_F. Specially at the transition to Mott insulating phase, the velocity of the spin-1 mode will be $2v_F$, i.e. the triplet mode (if we call it *triplon*) moves twice faster than the underlying Dirac electrons. This prediction can be tested not only in gapped graphene system, but also on a platform based on cold atoms engineered to mimic a massive Dirac theory at nano-Kelvin energy scales. The existence of a mode whose velocity can be twice the velocity of underlying Dirac fermions may have interesting implications in teleportation of quantum information, as well as in the spin-only forms of transport.

The above two modes discussed here exist for small values of U too. Therefore it should be possible to check for their experimental consequences. The well isolation of the energy scale ω_0 from the PHC edge ω_Δ makes the gapped graphene more interesting for performing neutron scattering experiments. In this work we find that the triplet excitation exists below the Mott transition. Moreover, the Mott insulating phase has its own magnetic excitations. Therefore in the gapped graphene we anticipate a triplet excitation over a large range of values of the Hubbard parameter U. Such low-energy spin-1 branch of excitations will have a characteristic T/v_s^2 contribution in the specific heat at constant volume, where v_s is the velocity of spin modes. Below the Mott transition, there will be another T/v_c^2 contribution coming from singlet charge modes. The influence of these modes on various properties of gapped graphene remains to be investigated.

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